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Absolute determinations of K-shell photoelectric cross-sections for uranium at the gamma-ray energies 1173 and 1332 keV

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With 1 figure in the text

Introduction

Recently, the authors of the present paper investigated a method for the beta-spectrometric determination of absolute values of internal conversion coefficients ε (Hultberg and Stockendal 1959, henceforth referred to as [HS]). An application was also made to the complex decays of Bi^{205} and Bi^{206} (Stockendal and Hultberg 1959, henceforth referred to as [SH]). Because ε is directly proportional to the photoelectric cross-section τ, according to Eq. (8) in [HS], the accuracy in the determination of ε will be directly influenced by the accuracy of τ. The latter quantity can be obtained from the tables of White (1957) for a number of elements. However, the tabulated photoelectric cross-sections could only be derived in a graphical way by joining calculations from several theories, each covering part of the energy range, with help of experimental values from absorption measurements (White 1957). Since the latter experiments cannot separate the contributions from the different electron shells the comparison with tabulated K-shell cross-sections τ_K implies a knowledge of τ_a/τ_K $(\tau_a = \tau_K + \tau_L + \cdots)$. *Experimentally*, the latter ratio is difficult to obtain, although it is expected to be rather insensitive to energy. *Theoretically*, the difficulties are considerable even for the K-shell, and they must be considered as almost insuperable if all shells are to be included, at least for heavy elements. A careful experimental study of τ_a/τ_K was reported by Hultberg 1959 (henceforth referred to as [H]), where the absorption ratios of the uranium atom were obtained in the beta-spectroscopically important energy region. The results of the latter investigation permit the conversion of τ_a to τ_K, and thus a direct comparison of absorption measurements with the tabulated cross-sections was made possible. For uranium it then turns out that the agreement with White's tables is satisfactory at the gamma-ray energies 412 and 662 keV (within 5 %). At 1332 keV, however, White gives $\tau_K = 5.5$ barns while $\tau_K = 4.7$ barns can be derived from the values given by Colgate 1952 (cf. Table 1 in [H]). Here the deviation from the tabulated value must be considered as being large (15 %), and it is far outside the experimental accuracy of 2 %, as claimed by Colgate. If the experimental value is right it would be serious for the theories underlying the tables, even if allowance is made for systematic errors in the diagrammatical fitting procdure. On the other hand, absorption measurements are always difficult to perform and since, above 1 MeV, τ_a is small compared with the sum of the cross-sections of the other mechanisms contributing to the absorption of gamma radiation, it is obvious that a reliable determination of τ_a from such experiments must be very difficult above 1 MeV.

In [HS] it was suggested that Eq. (8) also be used in the reverse sense, i.e. for a determination of absolute values of τ. The present paper is intended to report an application of this new technique to the 1173 and 1332 keV transitions in the decay of Co^{60}, in order to test the tabulated photoelectric cross-sections in the energy region where the above-mentioned discrepancy was found. Moreover, it is slightly above 1 MeV that the validity of Hulme's numerical calculations begins to break down while, on the other hand, such energies are too low for the Hall formula to be strictly applicable. It should then be of interest to test the tabulated τ_K in the critical energy region 1–1.5 MeV, where the Hulme and Hall theories were smoothly joined by White.

Experiments

The radioactivity used consisted of about 7 mC Co^{60}, obtained from the reactor at Chalk River, Canada, in the form of nitrate solution of high specific activity (15–20 C per gram). A thin and approximately circular source (diameter 7 mm) was prepared, using the electroplating technique. It was fixed onto an aluminium frame which was mounted on the sliding rod, shown in Fig. 3 in [H], so that the source took the correct position behind the converter ($\theta = 0$). The uranium converter was circular (diameter 14.6 mm) and had the thickness $d = 2.19 \pm 0.02$ mg/cm^2 U. It was fastened at the end of the vacuum-lock shaft, in the manner shown in Fig. 3 in [H]. From Fig. 3a) it is seen that the converter can be effectively removed from the spectrometer vacuum chamber by simply pulling out the shaft. The converter can then be said to take a "passive", or "outer", position, in contrast to the "active", or "inner", position in front of the source. The described arrangement was very convenient in that it permitted a rapid and easy switching between the measurements of conversion lines and photolines, by moving the converter between the outer and inner positions. As can be understood from Fig. 3a) of [H] the latter movement could be performed from outside the spectrometer, without disturbing the instrumental adjustments in any way. The distance a between the source and the converter (cf. Fig. 12 of [H]) was 2.0 mm. The resulting difference in the solid angle, under which the spectrometer entrance aperture is seen from a point on the converter and from a point on the source, is negligible, due to the small value of a/m. The quantity m is the distance from the point of emission to the aperture centre. Here we have $a/m \approx 1/160$.

For the 1173 and 1332 keV transitions in Ni^{60} conversion lines and uranium photolines were recorded, the converter taking the outer and inner positions, respectively. It should be pointed out that the construction of the vacuum lock device permits the inner position of the converter to be accurately reproduced (the exact position is, however, not too critical, cf. section H of [H]). All recordings were made using the scintillation equipment, described in [SH]. Three complete conversion line–photoline sets were recorded, apart from a preliminary run. A possible contribution, in the conversion line measurements, from photoelectric conversion in the source itself was estimated to be about 1 count/min which clearly is negligible. Fig. 1 illustrates one of the runs.

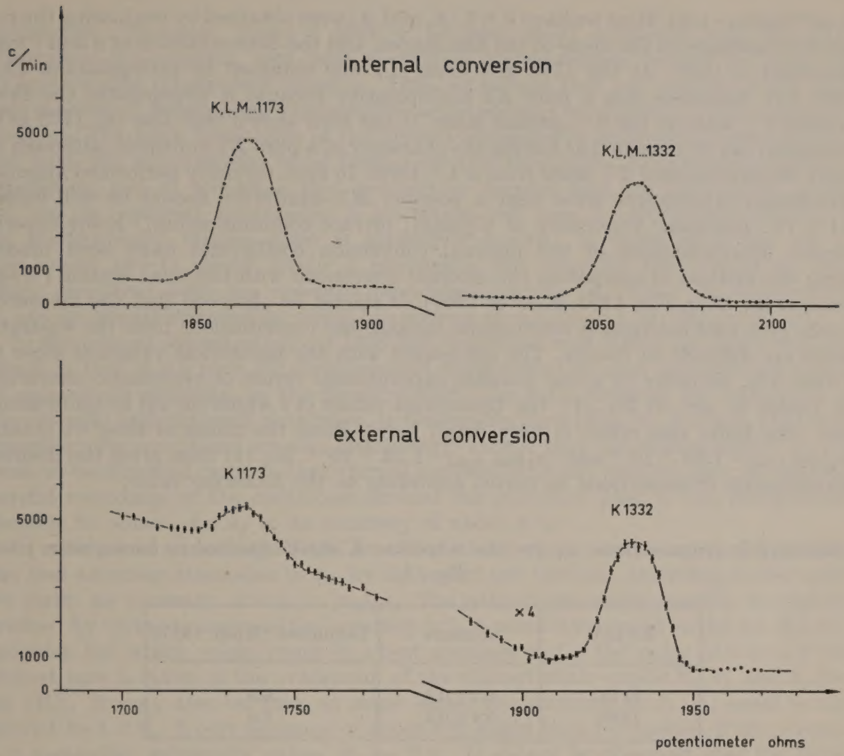

Fig. 1. Conversion lines and photolines from cascade transitions in Ni^{60}, of energies 1173 and 1332 keV. A 2.19 ± 0.02 mg/cm^2 circular U converter was used (radius 7.3 mm). It should be remarked that the areas of the line shapes were determined by considering recordings of the continuum, extending over a much larger energy region than that shown in the diagram. It is interesting to compare the ratio between the counting rates of internal and external conversion, obtained here, with that of Fig. 3 in [HS]. The same source was used for internal and external conversion ($k = 1$).

Results and conclusions

The *absolute* value τ of the photoelectric cross-section can be written, for a specific shell,

$$\tau = \frac{A_\gamma}{A_\beta}\,\frac{\varepsilon}{f\,d\,k\,b}, \tag{1}$$

according to Eq. (8) in [HS]. ε is the internal conversion coefficient of the transition in question, A_γ and A_β are the intensities of the photoline and the conversion line, respectively, f is a correction factor[1], d is the thickness of the converter material, k is the relative source strength, and b is a dimension factor $= 2.531 \times 10^{-6}$ atom ×

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[1] For the calculation of f the Co^{60} source can be regarded as a point source, since here we have $c_Q < \frac{1}{2} c$ (see section *H* of [H]).

$\times$ cm^2/(barns $\times$ mg). Here we have $k = 1$. A_γ and A_β were obtained by evaluating the respective averages of the areas of the line shapes, and the determination of d and f was described in [HS]. At the 1173 keV energy, f was obtained by interpolation. The 1332 keV transition has a pure $E2$ multipolarity because it depopulates the first excited $2+$ state to the $0+$ ground state. It has been shown that also the 1173 keV transition can be regarded as having the character of a pure $E2$ multipole, although it feeds the first excited $2+$ state from a $4+$ level. In fact, carefully performed angular correlation experiments show that a possible $M3$ admixture should be well below 0.1 % (E. Karlsson, University of Uppsala, private communication).[1] Some experimental determinations of the internal conversion coefficients have been made, using the method of comparing the internal conversion with the total feeding (Waggoner *et al.* 1950, Fan 1952, Keister 1954). It should be observed that the measurements give *total* conversion coefficients, because the contributions from the separate shells are difficult to resolve. The agreement with the theoretical values of Rose is within 5 %. In order to avoid possible experimental errors of systematic character we prefer to use, in Eq. (1), the theoretical values of ε which should be quite accurate (the finite size effect is very small here). From the tables of Rose we obtain $(\alpha_2)_{1173,\,\text{total}} = 1.65 \times 10^{-4}$ and $(\alpha_2)_{1332,\,\text{total}} = 1.25 \times 10^{-4}$. Eq. (1) then gives the desired photoelectric cross-sections in barns, according to the following table.

Photoelectric cross-sections τ_K for the uranium K-shell, obtained in barns/atom from Eq. (1).

$h\nu$ keV	Authors	Tabulated (White 1957)
1173	7.2 ± 0.5	6.9
1332	5.4 ± 0.3	5.5

The indicated errors are quadratically composed of the following contributions: $(A_\gamma/A_\beta)_{1173}$ 5 %, $(A_\gamma/A_\beta)_{1332}$ 3 %, ε 3 %, f 4 % and d 1 % (cf. discussion in [HS]). The choice of a 3 % uncertainty in ε represents a kind of "best guess", partly based on the systematic comparison of the Rose and Sliv values. The agreement between the latter calculations becomes increasingly better when Z decreases, due to the diminishing influence on ε from the nuclear finite size effect. It should be observed that our estimated uncertainty agrees with the deviation of available experimental data from the theoretical values.

From our results it is obvious that τ_K, as deduced from White's tables, satisfactorily represents the photoelectric cross-sections for the uranium K-shell in the critical energy region 1.1–1.4 MeV, to within 5 %. Thus, it seems probable that Colgate's τ_K-value at 1332 keV is too low.

To our knowledge the technique presented here has never before been used for the measurement of photoelectric cross-sections. The method has the advantage of giving a quantity that is directly obtained from the theoretical treatment of the photoelectric effect, namely the K-shell cross-section τ_K. In contrast, absorption measurements only give the cross-section τ_a for the whole atom, in an indirect way (cf. Introduction). Moreover, above 1 MeV τ_a becomes small compared with the total absorption cross-section, and here absorption measurements cannot be expected to give reliable information about τ_a. Thus we believe that our method is the only one capable of determining photoelectric cross-sections accurately at high quantum energies. The main limitations of the present technique lie in the difficulties associated with the requirements of a) finding suitable transitions, b) obtaining sufficient intensities, and c) preparing thin radioactive sources that are sufficiently strong for use in external conversion. A transition will here be considered *suitable* if its multipolarity mixing ratio is accurately known. In cases where the use of *one* source ($k=1$) would imply a conversion line intensity that would be very large, compared with the expected photoline intensity (cf. [HS]), the requirement c) can be largely released by using *two* sources ($k = k_\gamma / k_\beta > 1$, cf. [HS] and [SH]).

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[1] An $M3$ admixture of 0.1 % corresponds to an increase in ε of 0.3 % which is negligible.

In the present investigation the 1332 keV transition well fulfils the above requirements in that it is a pure $E2$ multipole and constitutes the most energetic transition of appreciable intensity, in the decay of Co^{60}. The ratio between the intensities of the internal and external conversion is favourable here (Fig. 1). For the 1173 keV transition the conditions are also satisfactory, except that the requirement of a good peak-to-background ratio, in the external conversion, is not fulfilled here. By making careful recordings of the continuum around the photoline peak it was nevertheless possible to obtain A_γ / A_β to an accuracy of about 5 %.

In conclusion, something should be added about what can be expected regarding the best accuracy attainable in τ_K, by the use of our method. According to the table we claim an accuracy of 6 % in $(\tau_K)_{1332}$. The latter figure might possibly be slightly lowered by trying to improve the accuracy in f. A possibility would be the use of a rectangular foil which might result in a best accuracy in f of the order of 1–3 %, if the utmost care is taken in the evaluation of the characteristic angles ϑ_0, ϑ_1, and ϑ_2 (see ref [H]). It may also be that, in some cases, the accuracy in A_γ / A_β could be improved to 1–2 %. A best accuracy of about 4 % might then be reached, if the error in ε is somewhat arbitrarily taken to be 3 %. It should be observed that the latter figure cannot be given generally, but has to be considered from case to case.

Note added in proof.—Experiments have very recently been performed by B. Johansson and R. Stockendal with the purpose to lower the Compton-background in external conversion measurements. A thin scintillator and a light-guide were placed between the converter and the source and mounted on a photomultiplier tube. Using anticoincidence techniques a decrease with, in some cases, 70 % of the background was observed, while the photoline intensity remained the same. The experiments are being continued and the results will be published later.

SUMMARY

Photoelectric K-shell cross-sections for uranium have been determined *absolutely* at the gamma-ray energies 1173 and 1332 keV, by the beta-spectrometric comparison of conversion lines and photolines. The results are $\tau_{K,1173} = 7.2 \pm 0.5$ barns and $\tau_{K,1332} = 5.4 \pm 0.3$ barns. These values agree with τ_K as inferred from the tables of White (NBS Circular No. 583, 1957), to within 5 %. The present paper is a report of the first application of a new technique for the determination of τ_K. Advantages and limitations of the method are discussed.

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